Polymer Sci., **18**, 1053 (1974).
Everage, A. E., and R. L. Ballman, "A Mechanism for Polymer Melt or Solution Fracture," *ibid.*, 933.
Finlayson, B. A., "The Galerkin Method Applied to Convective Instability Problems," *J. Fluid Mech.*, **33**, 201 (1968).
Gallagher, A. P., and A. Mercer, "On the Behavior of Small Disturbances in Plane Couette Flow with a Temperature Gradient," *Proc. Royal Soc.*, **A286**, 117 (1965).
Gavis, J., and R. L. Laurence, "Viscous Heating of a Power Law Liquid in Plane Flow," *Ind. Eng. Chem. Fundamentals*, **7**, 525 (1968).
Green, T., "Oscillating Convection in an Elasticoviscous Liquid," *Phys. Fluids*, **11**, 1410 (1968).
Han, C. D., "A Theoretical Study on Fiber Spinnability," *Rheol. Acta*, **9**, 355 (1970).
———, and R. R. Lamonte, "Studies on Melt Spinning I," *Trans. Soc. Rheol.*, **16**, 417 (1972).
Han, C. D., "Entrance Region Flow of Polymer Melts," *AiChE J.*, **17**, 1480 (1971).
———, and R. R. Lamonte, "Melt Flow Instabilities in Capillary Flow of Two Phase Polymer Systems," *ibid., Polymer Eng. Sci.*, **12**, 77 (1972).
———, "A Study of Polymer Flow Instabilities in Extrusion," *ibid.*, **11**, 385 (1971).
Huilgol, R. R., "Steady Temperature Fields in Second Order Fluids," *Acta Mech.*, **7**, 252 (1969).
Joseph, D. D., "Variable Viscosity Effects on the Flow and Stability of Flow in Channels and Pipes," *Phys. Fluids*, **7**, 1761 (1964).
———, "Stability of Frictionally Heated Flow," *ibid.*, **8**, 2195 (1965).
———, and W. H. Warner, "Parameter Value Excluded by Existence Conditions for Buoyant Dissipative Motions in Vertical Channels," *Quart. Appl. Math.*, **25**, 163 (1967).
Kearsley, E. A., "The Viscous Heating Correction for Viscometric Flows," *Trans. Soc. Rheol.*, **6**, 253 (1962).
Lebon, G., and G. Nguyen, "Hydrodynamic Stability by Variational Methods," *Intern. J. Heat Mass Trans.*, **17**, 655 (1974).
Marrucci, G., and R. E. Murch, "Steady Symmetric Sink Flows of Incompressible Simple Fluids," *Ind. Eng. Chem. Fundamentals*, **9**, 498 (1970).
Martin, B., "Some Analytic Solutions for Viscometric Flows of a Power Law Fluid with Heat generation and Temperature Dependent Viscosity," *Intern. J. Nonlinear Mech.*, **2**, 285 (1967).
———, "Theoretical Aspects of Screw Extruder Design," *Plastics and Polymers*, 113 (Apr., 1970).
Matovich, M. A., and J. R. A. Pearson, "Spinning a Molten Threadline," *Ind. Eng. Chem. Fundamentals*, **8**, 512 (1969).
McIntire, L. V., "On the Initiation of Melt Fracture," *J. Appl. Polymer Sci.*, **16**, 2901 (1972).
———, and W. R. Schowalter, "Stability of Viscoelastic Fluids," *Trans. Soc. Rheol.*, **14**, 585 (1970).
McIntire, L. V., "Use of Retarded Motion Expansions is Hydrodynamic Stability Analysis," *Physics Fluids*, **14**, 1074 (1971).
———, and W. R. Schowalter, "Hydrodynamic Stability of Viscoelastic Fluids," *AIChE J.*, **18**, 102 (1972).
Moore, C. A., and J. R. A., Pearson, "Experimental Investigations into an Isothermal Spinning Threadline," University of Cambridge, Polymer Processing Research Centre Report #14 (1972).
Nahme, R., "Beitrage zur hydrodyamischen theorie der Lagerreibung," *Ingr. Arch.*, **11**, 191 (1940).
Pearson, J. R. A., *Mechanical Principles of Polymer Melt Processing*, Pergamon Press, New York 77-119 (1966).
Porteous, K. C., and M. M. Denn, "Elastic Effects in Flow of Viscoelastic Liquids," *Chem. Eng. J.*, **2**, 280 (1971).
Rothenberger, R., D. H. McCoy, and M. M. Denn, "Flow Instability in Polymer Melt Extrusion," *Trans. Soc. Rheol.*, **17**, 259 (1973).
Sadd, M. H., "A Note on Shear Waves in BKZ Fluids," *ibid*, 647.
Spearot, J. A., and A. B. Metzner, "Isothermal Spinning of Molten Polyethylenes," *ibid.*, **16**, 495 (1972).
Stevenson, J. F., and R. B. Bird, "Elongational Viscosity of Nonlinear Elastic Dumbell Suspensions," *ibid.*, **15**, 135 (1971).
Stevenson, J. F., "Elongational Flow of Polymer Melts," *AIChE J.*, **18**, 540 (1972).
Sukanek, P. C., "Poiseuille Flow of a Power Law Fluid with Viscous Heating," *Chem. Eng. Sci.*, **26**, 1775 (1971).
Sukanek, P. C., C. A. Goldstein, and R. L. Laurence, "The Stability of Plane Couette Flow with Viscous Heating," *J. Fluid Mech.*, **57**, 651 (1973).
Sukanek, P. C., and R. L. Laurence, "An Experimental Investigation of Viscous Heating in Some Simple Shear Flows," *AIChE J.*, **20**, 474 (1974).
Tanner, R. I., "Network Rupture and the Flow of Concentrated Polymer Solutions," *ibid.*, **15**, 177 (1969).
Tordella, J. P., "Capillary Flow of Molten Polyethylene—A Photographic Study of Melt Fracture," *Trans. Soc. Rheol.*, **1**, 203 (1957).
———, "Unstable Flow of Molten Polymers: A Second Site of Melt Fracture," *J. App. Polymer Sci.*, **7**, 215 (1963).
Trowbridge, E. A., and J. H. Karran, "A Discussion of Critical Parameters which can Occur in Frictionally Heated Non-Newtonian Fluid Flows," *Intern. J. Heat Mass Transfer*, **16**, 1833 (1973).
Turian, R. M., "The Critical Stress in Frictionally Heated Non-Newtonian Plane Couette Flow," *Chem. Eng. Sci.*, **24**, 1581 (1969).
Turian, R. M., "Viscous Heating in the Cone and Plate Viscometer," *Chem. Eng. Sci.*, **20**, 771 (1965).
Vest, C. M., and V. S. Arpaci, "Overstability of a Viscoelastic Fluid Layer Heated from Below," *J. Fluid Mech.*, **36**, 613 (1969).
Vinogradov, G. V., N. I. Insarova, B. B. Boiko, and E. K. Borisenkora, "Critical Regimes of Shear in Linear Polymers," *Polymer Eng. Sci.*, **12**, 323 (1972).
Vlachopoulos, J., and M. Alam, "Critical Stress and Recoverable Shear for Polymer Melt Fracture," *ibid.*, **12**, 184 (1972).
Zamodits, H. J., and J. R. A. Pearson, "Flow of Polymer Melts in Extruders," *Trans. Soc. Rheol.*, **13**, 357 (1969).

Manuscript received February 11, 1975; revision received May 29, and accepted May 30, 1975.

Phase Separation of Primary Dispersions in Beds Packed with Spherical Packings

Studies have been made of the behavior of drops in primary dispersions flowing through beds packed with glass ballotini in order to establish the mechanism of phase separation. The buoyancy and surface forces have been analyzed in terms of drop size and shape in the interstice in the packing and the physical properties of the dispersion. A mathematical model has been developed to describe drop behavior in the bed, and the drop size in the effluent dispersion has been predicted from established correlations.

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SCOPE

Although packings are used extensively as an aid to separate the phases of primary dispersions, particularly when the density difference between the phases is small, little is known about either the behavior of the dispersed phase in the packing or the parameters that regulate the size of the drops which emerge from the packed bed. This is surprising because the effluent drop size ultimately controls the success of the phase separation. Considerable work has been published on the qualitative aspects of packings for the phase separation of both primary and secondary dispersions, and it has been established that wetting characteristics and surface roughness are important features. However, very little work has been published on the mechanism of drop coalescence and phase separation in packed beds. Therefore, in order to characterize the packing, phase separation studies have been undertaken using single nozzles with a layer of spherical packing and with beds packed with glass ballotini. In addition, the forces acting on a drop being squeezed through the interstices between packing elements have been analyzed to establish criteria for the selection of a packing to separate primary dispersions. Finally, the parameters controlling the drop size in the effluent dispersion have been investigated experimentally, and an attempt has been made to predict the effluent drop size from the properties of the packing and the system undergoing phase separation.

CONCLUSIONS AND SIGNIFICANCE

Coalescence and phase separation of primary dispersions in packed beds depend on a drop retention mechanism in the bed and a drop release mechanism from the exit layers of the packing. A two-dimensional mathematical model has been proposed to predict the upper and lower limits of size of a drop that would be retained by a given packing. These sizes have been confirmed experimentally with single nozzles, with a layer of ballotini packing, and by treating primary dispersions in beds packed with spherical packings. The model has been presented in graphical form in terms of drop size and packing interstice size with the group $[\Delta\rho g/2\gamma]$ characterizing the physical properties of the system as parameter. This chart can be used as the basis for the design of a packing to separate the phases in a primary dispersion.

Statistical treatment of the experimental results from the phase separation of primary dispersions in beds packed with glass ballotini showed that the depth of packing in the bed, the superficial velocity of the dispersed phase, preferential surface treatment, and inlet drop size had only minor effects on the drop size in the effluent dispersion which emerged from the packing. The effluent drop size depends to a great extent on the size of the packing elements in the exit layers of the bed; these should be as large as possible but must be compatible with the limits predicted by the drop retention chart. The size of the drop in the effluent dispersion can be estimated, albeit approximately, from the equivalent void diameter of the packing and correlations of Meister and Scheele (1967) for discharge of a liquid from circular nozzles.

In many industrially important phase separation processes the rate of droplet coalescence is so slow that some form of coalescing aid must be introduced to maintain the throughput of the plant. The particular coalescing aid employed depends on the characteristics of the dispersion; that is, whether it is a primary or a secondary dispersion. For the very stable secondary dispersions, the rate of phase separation can be increased by adding electrolytes, de-emulsifying agents, or finely divided adsorbent gases or solids to increase the coalescence and/or settling rates. Frequently this type of treatment must be followed by further operations to remove the separating agent, and therefore some form of mechanical technique is generally preferred. This may be a packing or membrane, centrifuge, hydrocyclone, or an electrostatic coalescer. Packings are the most economical and are utilized to induce and accelerate phase separation of both primary and secondary dispersions. However, the type of packing required for a given duty depends on the characteristics of the dispersion. For the treatment of a primary dispersion, the packing should be preferentially wetted by the dispersed phase, and numerous materials are available ranging from metal swarf, pebbles, conventional ring, and saddle packings to knitted and woven meshes. The important factors that control the performance of these packings are the materials of construction and the nature of the surface of the packing, the voidage and the size of the packing element, or the stitch if knitted or woven packings are used. For the treatment of secondary dispersions, the wetting properties of the material are less important (Sareen et al., 1966). The drops are less than the equilibrium size (Padday, 1957) so that they will be very nearly spherical and will tend to make only point contact with the coalescing surface. This has been confirmed by Kintner (Davies and Jeffreys, 1969) who showed that surface roughness was more important than the wetting properties of the packing. Furthermore, fibrous beds of cotton, glass wool, and metal and polymer strands have been used for the separation of secondary dispersions irrespective of which phase was dispersed. Generally the drop size in these dispersions is less than 10μ, while the minimum fiber pitch of most of these packings is of the order of 30μ (Shalhoub, 1975), so that the secondary dispersion entering the bed flows freely between the packing elements. Initially the drops are attached to the fibers by impaction and adhesion, as described by Spielman et al. (1972, 1973), and this is followed by interdrop coalescence occurring repeatedly with these attached drops until the enlarged drops are displaced from the packing site by the drag forces. This invariably occurs at drop sizes of the order of 1.0 mm, very much larger than the space between the fibers, so that the behavior of the drops near the discharge section of the bed will be similar to that in packings treating primary dispersions. Therefore, for all phase separation processes in packed beds, the interstices between the packing elements must be small enough to accommodate the range of drop sizes encountered in the dispersion and yet maintain the pressure drop as low as possible. It is well known that when the drop size in a primary dispersion is less than that of the interstice in the packing, the coalescence rate is very low, even when the dispersed phase wets the packing. This is due to the low adhesion force, compared with the other forces, acting on the relatively large drop. Further-

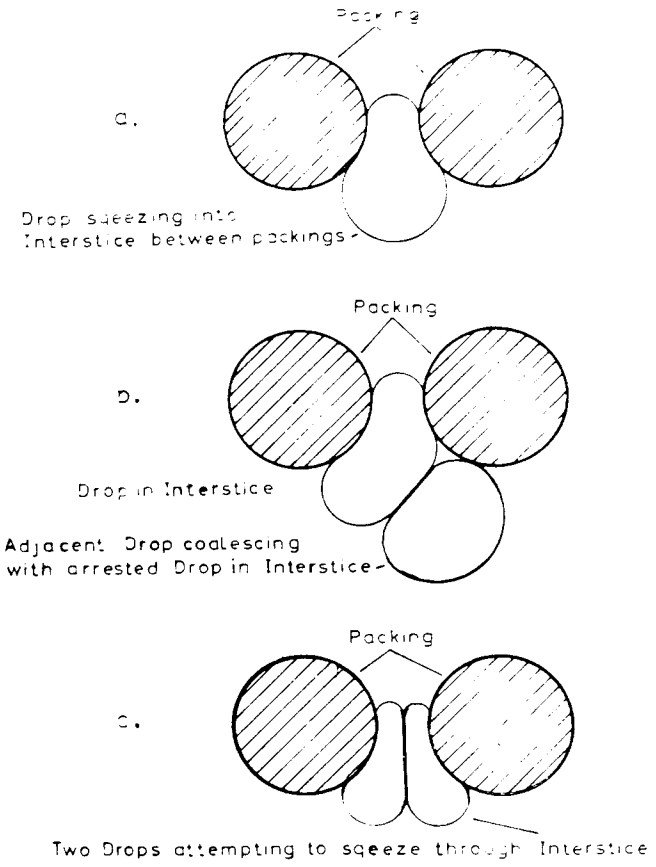


Fig. 1. Drop behavior in spherical packings

more, drops of the size found in a primary dispersion are elastic and tend to bounce off the strand of packing rather than adhere. Consequently, in order to ensure that coalescence occurs in the packing, irrespective of whether the dispersed phase wets the packing, the drop size in the entering dispersion must be larger than the interstice. When these conditions exist, the situation will be as represented in Figure 1. That is, the drop will be trapped, at least momentarily, and must undergo distortion in order to pass through the space in the packing. If it resists distortion, the drop will be retained in the aperture until either the film of the continuous phase between it and the packing ruptures or, as other drops in the dispersion will soon accumulate around the stationary drop, until interdrop coalescence occurs. Repeated interdrop coalescence will be necessary to increase the size of the retained drop sufficiently to provide the buoyancy to force the drop through the aperture in the packing, and, unless the dispersed phase wets the packing, it will most probably flood. The range of drop sizes that would be retained in a packing has not been studied hitherto, although Davies and Jeffreys (1969) have shown that when the ratio of the aperture diameter to drop diameter is less than 0.4, the drop will not pass through the space. Therefore, to identify the parameters controlling droplet coalescence behavior in packed beds, an investigation was undertaken, and in order to specify the geometry of the packing, it was decided to utilize spherical packing elements in the bed. Spherical packing elements can be arranged in a body centered or face centered configuration, and drops attempting to pass through the interstices between the spheres will be distorted in a vertical and horizontal plane. Therefore, the analysis of the behavior of the dispersion in such a packing should realistically be based on a three-dimensional model. However, since the distortions in the

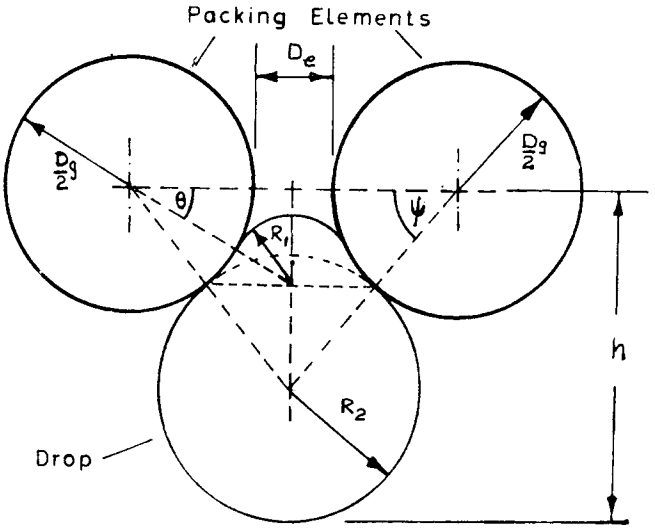


Fig. 2. Model of drop residing in packing interstice.

horizontal plane will be small compared with those in the vertical plane, a two-dimensional model has been developed and is presented below.

THE MODEL

Consider a dispersion in which the dispersed phase has a lower density than the continuous phase so that the drops will rise through the continuous phase, and let a drop of diameter D_b reside temporarily in the interstice between two elements of spherical packing as shown in Figure 2. The buoyancy forces will tend to force the drop through the void, and this will distort the drop as illustrated. The surface forces will resist distortion, and, provided they exceed the buoyancy forces, the distortion will be insufficient for the drop to pass through the aperture between the packings. Adjacent drops will not make a great contribution to the buoyancy force of the retained drop, since their buoyancy forces will be dissipated by their own distortion. However, when the retained drop coalesces with an adjacent drop, the buoyancy force is doubled, whereas the surface forces of the resulting drop are only slightly changed. It will undergo further distortion and, after a number of interdrop coalescences, will become sufficiently distorted to be forced through the aperture between the packing elements. At this point the buoyancy force just exceeds the surface forces, and, provided the voidage in the packing is uniform throughout the bed, the enlarged drop will then pass through the remainder of the packing unimpeded. Nevertheless its flow rate will be low so that viscous drag forces will be small and can be neglected. Therefore, an estimate of the maximum size of a drop that will be retained in the interstice between packing elements can be made by equating the buoyancy and surface forces on a drop attempting to pass through the aperture in the packing. Then, a force balance on the drop shown in Figure 2, by using the Laplace equation, leads to

$$\frac{h\Delta\rho g}{2\gamma} = \frac{1}{R_1} - \frac{1}{R_2} \qquad (1)$$

The radius of the leading surface of the drop R_1 and the receding surface R_2 will vary with the distortion of the drop in the aperture. However, for any situation it can be shown from simple geometrical considerations (Wilkinson, 1974) that the radius of the leading surface of the drop is

$$R_1 = \frac{D_g(1 - \cos\theta) + D_e}{2\cos\theta} \quad (2)$$

and that of the lower surface of the drop is

$$R_2 = \frac{D_g(1 - \cos\psi) + D_e}{2\cos\psi} \quad (3)$$

Also, from geometrical considerations in Figure 2

$$h = R_2 + \frac{(D_g + D_e)\tan\psi}{2} - \frac{1}{2}[(D_g + D_e)(\tan\theta - \sec\theta) - D_g] \quad (4)$$

Equations (2), (3), and (4) can be combined with the initial volume of the drop of diameter D_b, and in terms of the dimensions in Figure 2

$$D_b^3 = R_2^3(2 + 2\sin\psi - \sin\psi \cdot \cos^2\psi) + D_g(\sin\psi - \sin\theta)(R_1^2\cos^2\theta + R_1R_2\cos\theta\cos\psi + R_2^2\cos^2\psi) + 2R_1^3(1 - \sin\theta)(\cos^2\theta - \sin\theta + 1) \quad (5)$$

The basic pressure balance is expressed by Equation (1), and this can be solved in terms of Equations (2), (3), (4), and (5) since ψ and θ can be expressed in terms of the packing element diameter D_g and the equivalent diameter of the interstice D_e. The limiting condition is $(\theta = 0)$, since this represents the situation that the radius of the leading surface is equal to the radius of the aperture between the packing elements and the condition that the surface forces opposing the buoyancy forces are a maximum. Any further penetration of the drop into the aperture will result in a decrease in the resultant force, and the drop will continue to flow through the space in the packing by virtue of the dominant buoyancy forces. Equations (1) to (5) were compiled into an appropriate computer program to evaluate the resultant force E on the drop (Wilkinson, 1974). If E is positive, the drop will pass through; if it is negative, it will be retained. Typical results for a packed bed made up of spherical particles 0.6 m diameter with an aperture equivalent diameter of 0.248 cm between the particles is presented in Figure 3.

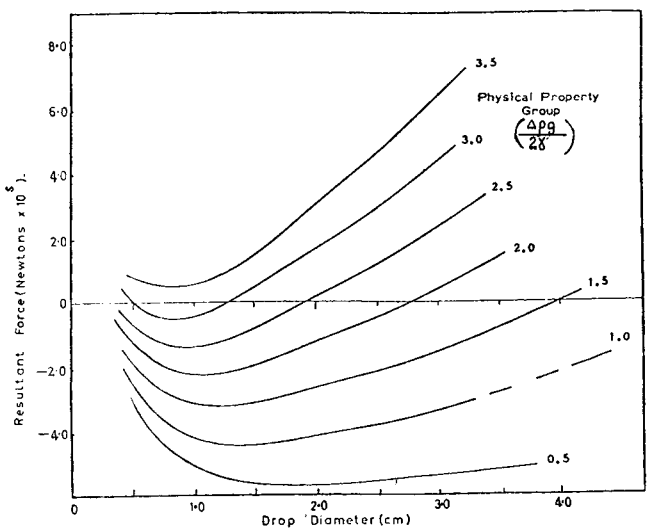


Fig. 3. Range of drop retention in spherical packings (packing dia 0.6 cm).

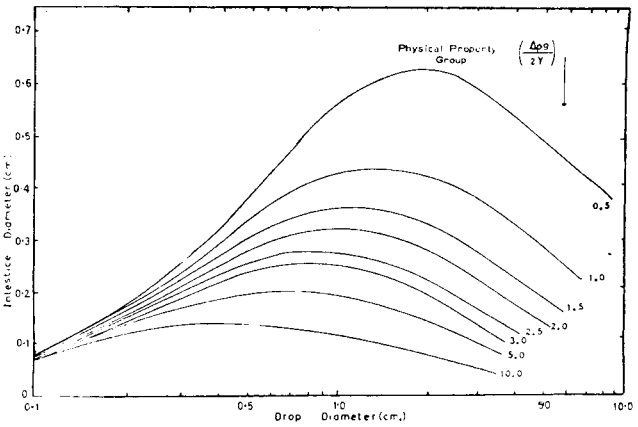


Fig. 4. Relation between size of drop retained and size of interstice.

In this figure the physical property group $[\Delta\rho g/2\gamma]$ is the parameter, and the horizontal line at $(E = 0)$ is the demarcation between droplet flow-through or retention in the interstice. For example, drops in a system with a physical property group 3.0 would be retained in the size range 0.5 to 1.25 cm, whereas when the physical property group is 3.25 or greater, no drops would be retained irrespective of their size. The computer program results for all the packing sizes and arrangements and the various drop sizes were analyzed to obtain the range of drop sizes that would be retained by different spherical packings. These results are presented in Figure 4. In this figure, void equivalent diameter is plotted against the drop diameter with the physical property group as parameter. The area beneath any curve corresponds to the region of drop sizes that would be retained in the interstices in a packing. Similarly, the space above any curve corresponds to the region where drops would flow through the packing unimpeded. For example, drops in a dispersion with physical properties corresponding to a physical property group of 3.0 flowing through a bed of spherical packing with interstice diameter 0.2 cm would be retained in the size range 0.5 to 1.5 cm as illustrated in Figure 4. This figure provides a basis for the design of a packing for a packed bed coalescer treating primary dispersions. The predictions will also be applicable to the exit regions of beds treating secondary dispersions, although the scale will be an order of magnitude lower. In conventional packings, such as knitted meshes, the geometries of the apertures are, of course, less well defined, and their axes are not always vertical. However, while spherical packing elements are rare in practice, this model provides a reasonable basis for the prediction of dispersion behavior in porous media and is believed to be capable of extension to conventional beds.

EXPERIMENTAL

The analyses presented above were tested by coalescing dispersions in beds packed with glass ballotini and by single nozzle experiments with a layer of glass spheres located in a specific packing arrangement immediately above the nozzle.

The apparatus shown in Figure 5 was utilized to study drop behavior in the aperture between packing elements. The aperture in the bed was simulated by attaching a ballotini to a glass rod with epoxy resin adhesive and arranging three or four of these spheres into a packing unit as shown in Figure 5. The ballotini could be arranged to resemble a cubic or triangular configuration as desired. The glass nozzle, which was interchangeable, was connected to a syringe pump via a glass tube as shown in the diagram, and the drop flow rate was controlled in the range 0.016 to 0.28 ml/s. Drops formed at the nozzle were allowed to rise into the space between the ballotini. If the drop was retained by the packing, the nozzle was replaced by

TABLE 1. COMPARISON OF PREDICTED AND EXPERIMENTAL RESULTS FOR DROP SIZE LIMITS FOR SINGLE NOZZLES

System	Physical property no. $(cm)^{-2}$	d_B (cm)	d_{vc} (cm)	Cubic Lower limit Exp (cm)	Cubic Lower limit Theory (cm)	Cubic Upper limit Exp (cm)	Cubic Upper limit Theory (cm)	d_{vt} (cm)	Triangular Lower limit Exp (cm)	Triangular Lower limit Theory (cm)
Diethyl carbonate-water	1.1	1.2	0.5	(0.72-NH)	NH	1.94	NH	0.22	0.3	0.25
		0.9	0.375	0.65	0.63	2.35	3.0	0.15	0.2	0.22
		0.6	0.26	0.40	0.32	4.1	6.5	0.12	0.2	0.15
Toluene-water	2.05	1.2	0.5	NH	NH	NH	NH	0.19	0.20	0.24
		0.9	0.375	(0.65-NH)	NH	1.9	NH	0.15	0.20	0.18
		0.6	0.25	(0.38-0.50)	0.45	2.5	2.6	0.11	0.20	0.14
Iso-octane water	2.9	1.2	0.5	NH	NH	NH	NH	0.20	0.34	0.28
		0.9	0.4	(0.8-NH)	NH	1.6	NH	0.14	0.25	0.17
		0.6	0.25	(0.45-0.55)	0.53	1.4	1.0	0.12	0.25	0.15
MIBK-water	11.1	1.2	0.5	NH	NH	NH	NH	0.19	NH	NH
		0.9	0.4	NH	NH	NH	NH	0.15	0.25	NH
		0.6	0.28	NH	NH	NH	NH	0.11	0.23	NH

(NH = no holdup)

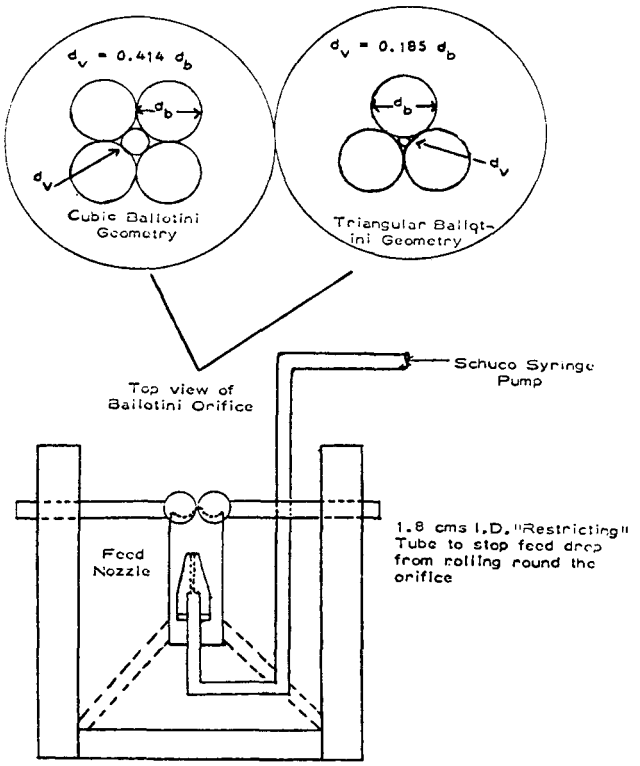
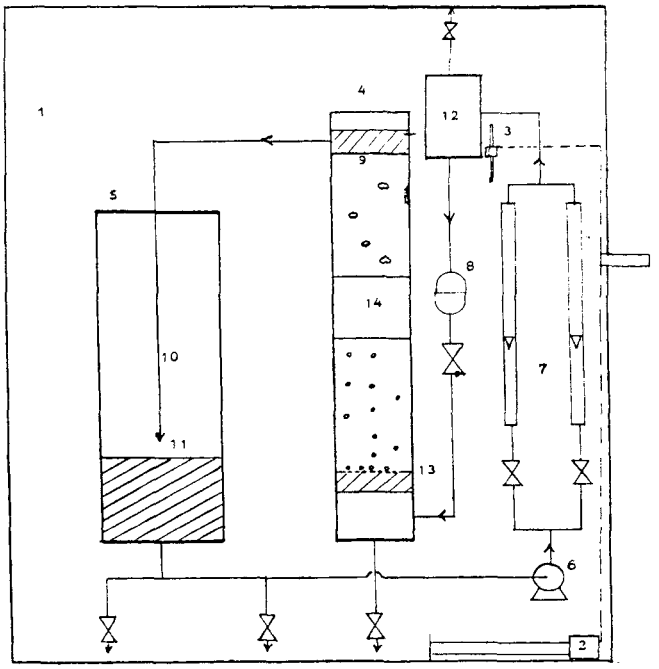


Fig. 5. Apparatus to estimate range of drop size retained by packing interstices.



1. Cabinet
2. Heat Exchanger
3. Capacitance Unit
4. 15 cm column
5. Organic Reservoir
6. Centrifugal Pump
7. Rotameters
8. Filter
9. Organic Water Interface
10. Overflow Pipe
11. Organic Air Interface
12. Head Chamber
13. Brass Distributor Plate
14. Ballotini Packing
15. Fl-Monitor Control Module

Fig. 6. Flow diagram of packed bed coalescer.

one of slightly smaller diameter, and this procedure was repeated until drops were produced which just passed through the interstice in the packing. Alternatively, if the drops produced initially passed through the aperture, drop sizes were increased by replacing the nozzles until drops were produced which were just retained by the ballotini. In this way the lower limit drop size was estimated. Each determination was repeated five times for different ballotini diameters and configurations, and the results obtained for the different systems studied are compared with the predicted drop sizes in Table 1.

The upper limit drop size was also estimated in the apparatus shown in Figure 5 by allowing drops to accumulate beneath and coalesce with the drop arrested in the aperture of the packing unit. As the drops accumulated beneath the packing unit they were prevented from dispersing and rolling around the outside of the packing unit by the restriction tube shown in Figure 5. Passage through the aperture occurred very abruptly after the accumulated drops had coalesced sufficiently for the buoyancy of the resultant drop to force it through the aperture. Therefore, the flow of the enlarged drop through the orifice between the packings was recorded by high-speed cine photography, and the upper limit drop size was estimated from photographs. The systems studied were the same as those used in the lower limit size estimations, and the results obtained are reported in Table 1.

Phase separation rates of dispersions were studied in the apparatus illustrated in Figure 6 which is self-explanatory and requires little comment. The packed bed coalescer was constructed from a glass column 15.0 cm nominal bore and 100 cm long. A brass distributor was fitted into one end of the column, and a glass packing support plate was located 45.0 cm from the distributor to ensure that drops in the dispersion attained their terminal velocity before entering the packing. The ballotini packing of uniform size was placed on the support plate by inserting a long stemmed funnel into an opening in the top of the column and then by adding the packing through

this with the column full of liquid. In this way a bed of the desired configuration and depth could be assembled without disconnecting parts of the equipment. The dispersed phase reservoir was of glass and had a capacity of 20.0 liters. It was connected to the coalescer via the rotameters by 1.0 cm bore glass piping. The whole apparatus was enclosed in a thermostatically controlled cabinet.

The packing to be inserted into the column was subjected to the preferential wetting treatment devised by Thomas (1971) to ensure that all the packing had a uniformly reproducible surface. The ballotini was first soaked in chromic acid for 24 hr and then thoroughly washed with distilled water, after which it was dried for 8 hr at above 150°C. The treated ballotini was immersed in the dispersed phase which would thereafter preferentially wet the ballotini surface irrespective of the surface free energy.

Experiments were carried out with packing of uniform size in the range 2.0 to 5.0 mm in beds varying in depth between 5.0 and 25.0 cm. During an experiment the coalescer was almost completely filled with the continuous phase. A dispersion, of uniform size drops, was then produced at the distributor which passed into the packing. Interdrop coalescence occurred, and when steady state conditions had been attained, the dispersed phase flow rate was recorded and photographs were taken of the dispersion entering and leaving the packing. The mean drop size was estimated from the photographs by using the Quantimet automatic analyzer. Details of the photographic techniques and of the analyzer are given by Wilkinson (1974). Experiments were carried out at different dispersed phase flow rates and initial drop sizes with various packing depths and ballotini sizes, and the results obtained are presented below.

DISCUSSION OF RESULTS

The results obtained for the single nozzle experiments are presented in Table 1. There it will be seen that there is good agreement between the predicted lower limit size of the retained drop and the experimental size for the triangular packing configuration and the smaller apertures in cubic arrangement. The agreement between the predicted and experimental lower limit drop size was less satisfactory for the larger drops, where in some instances no retention was predicted but in practice the drops were held up. In fact, the effective surface forces are smaller on the larger drops so that greater deformation is required for penetration and passage. In addition, slight variations were also observed in drop sizes emerging from the nozzle even though every care was taken to eliminate this effect.

The upper limit drop size, or breakthrough size, was very difficult to estimate accurately because breakthrough occurred very rapidly and, on a number of occasions, could not be analyzed, even with the aid of high-speed photography. It may be that inertial forces, associated with the increased flow rates of these larger drops, were becoming significant and producing some slurp penetration effects. When passage occurred immediately after a drop coalescence, the enlarged drop suddenly accelerated through the aperture to emerge as a jet which subsequently broke up into smaller drops. Furthermore, as the rear of the elongated drop emerged from the aperture, it tended to draw some of the inlet drops through the space between the packing elements. Hence it was not possible to estimate the upper limit drop size from the sum of the drops existing above the packing immediately after breakthrough. This effect was most pronounced with the small apertures between the ballotini arranged in a triangular configuration, and for this reason upper limit drop size results have not been reported for this configuration. However, generally the upper limit drop sizes were found to be lower than the predicted values, indicating a reverse trend to that expected from consideration of drag forces. The very rapid acceleration of the drop through the space between the packing elements and the breakup of the resulting jet would not occur within a packed bed because the packing

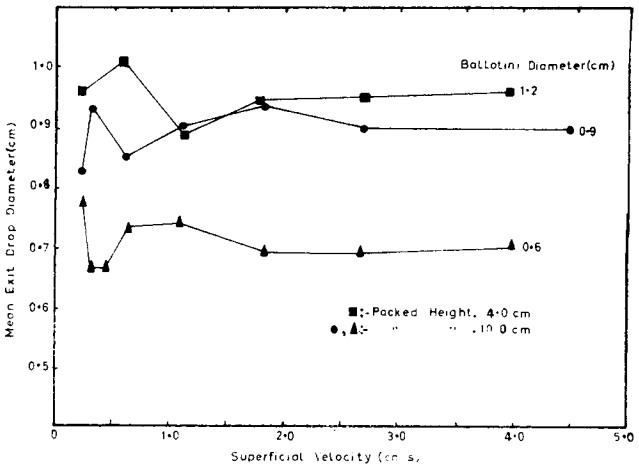


Fig. 7. Variation in exit drop diameter with superficial velocity.

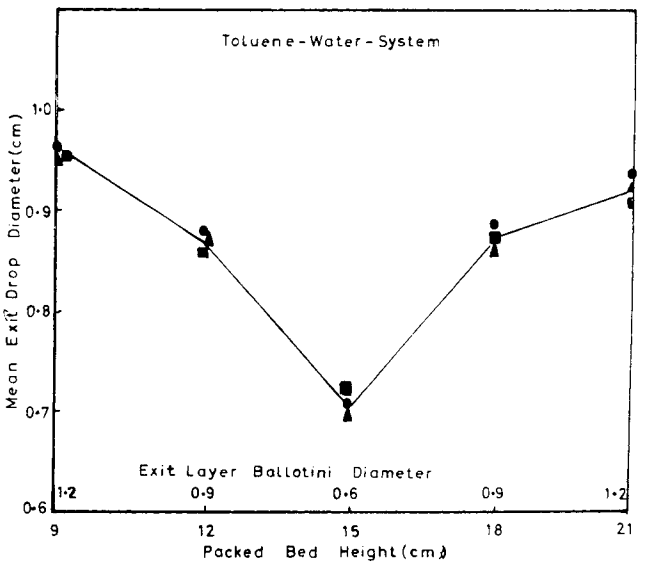


Fig. 8. Variation of exit drop diameter with exit layer ballotini diameter.

beyond the retention-coalescence point would dissipate the kinetic energy acquired by the drop. However, the mechanism described above would be expected to apply to drops emerging from a packed bed, suggesting that there are two distinct processes controlling phase separation and drop growth in a packed bed coalescer.

The drop behavior in passing through a single layer of packing was compared with the behavior of a dispersion in the 15.0 cm diameter bed packed with glass ballotini. The effects of dispersed phase flow rate, packing height, inlet drop, and ballotini diameter on the exit drop size were investigated, and a typical set of results are presented in Figure 7. This figure shows that ballotini size has a significant effect on the drop size in the dispersion leaving a packed bed. This is to be expected if the exit drops are formed at a series of orifices in the exit layer of the packing. The increase in the exit drop size with the increase in ballotini diameter follows from the fact that the mean void diameter increases as the particle size increases. This effect was further investigated by placing a layer of ballotini 1.2, 0.9, or 0.6 cm diameter on the discharge side of the packed bed, and the results obtained for different bed depths and packing sizes in the bed are summarized in Figure 8. These results confirm that the drop size in the effluent dispersion leaving the packing depends to a very great extent on the void size in the

exit layers of the packing. This factor is of great importance in the design of packed bed coalescers since, providing drop retention and coalescence occurs within the bed, it is desirable to install a layer of packing at the exit with as large a space between the particles as possible. The criteria specified in Figure 4 must however apply to this layer. The results reported in Figure 7 also show that the dispersed phase flow rate affects the phase separation efficiency of packed bed coalescers, but the behavior pattern is complex, depending on the properties and the geometry of the packing and on the retention-release mechanism in the packing. At low flow rates Figure 7 shows that the exit drop size varies considerably with flow rate. During these experiments it was observed that drops emerged from only a small number of points. Thomas (1971) referred to this as channeling and suggested that it might be related to preferential wetting of some of the packing by the dispersed phase. Duplicate experiments were carried out with nonwetted ballotini and with the same ballotini treated by the method described above to make it wettable, but no significant difference in performance was noticed. This suggests that preferentially wetted channels have little effect on the exit drop release mechanism. Alternatively the active exit release sites were thought to be related to local fluctuations in the aperture size between packing elements, but these active sites were observed to be distributed randomly over the exit of the bed. If the space between the packings were the dominant factor, it would be expected that the majority of the active exit release sites would be located near the column walls, since Ridgway and Tarbuck (1967) found a high voidage region within two particle diameters of the wall. It must therefore be concluded that, although the exit layers of the packing affect and largely control the exit drop size in the dispersion, the release sites also depend on flow within the packing. Nevertheless, examination of Figure 7 shows that irrespective of ballotini size the largest drops are released at low flow rates, and these were found to emerge from a small number of sites. Since it has been shown that generally exit drop size is related to void size, it is reasonable to assume that the initial active release sites are slightly larger than the mean size. As the flow rate is increased, the number of release sites increases and, since these are slightly smaller, the mean drop size in the effluent dispersion is also smaller. Continued increase in the dispersed phase flow rate results in the accumulation of drops in the form of a band at the entrance to the packing. Retention and most of the drop growth occurs in the entrance regions of the bed, but the packing continues to operate without flooding. The packing is analogous to a series of tortuous tubes with nozzles, and the exit drop size changes very little with velocity, as would be expected from the work of Meister and Scheele (1967) on drop formation from cylindrical nozzles in liquid-liquid systems.

Finally, all the experimental results were tested statistically and confirmed that the depth of packing in the bed, superficial velocity of the dispersed phase, preferential surface treatment, and inlet drop size had only small effects on the exit drop size in the effluent dispersion. The effluent drop size, however, depended to a great extent on the size of the ballotini packing and configuration in the exit layers of the bed. Therefore, an attempt was made to characterize the effluent drop size with the ballotini size in the exit layers of the packing through a drop-void ratio ϕ expressed as

$$\phi = \left[\frac{\text{Mean Drop Diameter in Effluent Dispersion}}{\text{Equivalent Void Diameter in Exit Layers of Packing}} \right]$$

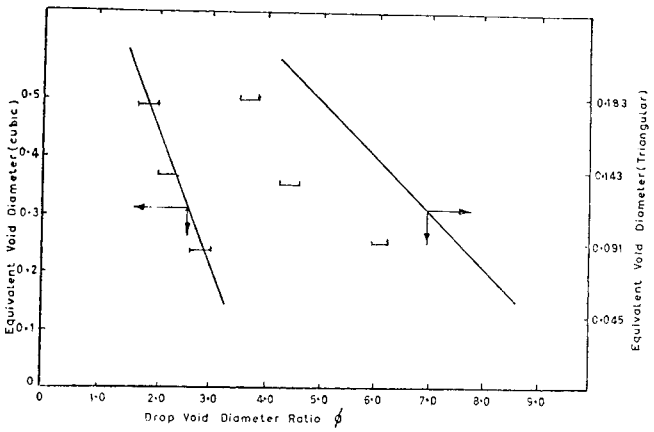


Fig. 9. Comparison of experimental and predicted drop void diameter ratios.

The equivalent void diameter is related to the packing size and configuration; thus for a cubic configuration, the equivalent cubic void diameter d_{vc} is related to the ballotini size d_b by $(d_{vc} = 0.414 d_b)$, while the equivalent triangular diameter d_{vT} is related to the ballotini size by $(d_{vT} = 0.155\ d_b)$.

The mean value of the drop-void diameter ratio was obtained for each experiment and plotted against the respective packing equivalent void diameter as shown in Figure 9, where it can be seen that there is a good linear relation between ϕ and d_{vc} or d_{vT}. These results were compared with predicted values of the drop-void diameter ratio by using the correlations of Meister and Scheele (1967). Since the velocity of effluent drop formation could not be evaluated, each drop-void diameter ratio was calculated over the velocity range 2.0 to 10.0 cm/s, that is, in the subjetting region, because jets were not observed in the experiments. The results obtained in Figure 9 are presented throughout over this range. It can be seen from Figure 9 that there is very good agreement between the experimental and predicted values of the drop-void diameter ratio for the cubic packing configuration, but the comparison is less satisfactory for the triangular configuration. This is believed to be due to difficulties in constructing a triangular packing configuration in the exit layers of the packing and also to the inability to define a circle through which the dispersed phase flowed that would be equivalent to the nozzle diameter in the Meister and Scheele correlation, since the slope of both the experimental and predicted plots for the triangular configuration in Figure 9 are the same. Therefore, it may be concluded that the drop size in the effluent dispersion leaving a packed bed is related to the diameter of the interstice between the packing elements. This is believed to be the principal mechanism irrespective of whether or not the dispersed phase wets the packing, since the flow of dispersed phase would fill in exit region interstices.

NOTATION

d_b = diameter of ballotini
d_v = equivalent diameter of interstice
d_{vc} = equivalent diameter of interstice–cubic configuration
d_{vT} = equivalent diameter of interstice–triangular configuration

D_b = drop diameter
D_g = diameter of ballotini
D_e = diameter of restriction in two-dimensional model
E = net force on drop
g = gravitational acceleration
h = differential head on drop giving buoyancy force
R_1 = radius of leading surface of distorted drop
R_2 = radius of lower surface of distorted drop

Greek Letters

γ = interfacial tension
$\Delta\rho$ = difference in density between phases
ϕ = drop-void diameter ratio
θ = angle between packing elements and spherical cap of drop
ψ = angle between packing element and spherical base of drop

LITERATURE CITED

Davies, G. A., and G. V. Jeffreys, "Coalescence of Droplets in Packings-Factors affecting the Separation of Droplet Dispersions," Filtration and Separation, 349 (1969).

Meister, B. J., and G. F. Scheele, "Generalized Solution of the Tomotika Stability Analysis for a Cylindrical Jet," *AIChE J.*, **13**, 682 (1967).

Padday, J. F., "A New Method for Measuring the Spreading Coefficient of a Liquid on a Solid Surface," in *Proc. 2nd Int. Cong. Surface Activity*, Vol. 3, Butterworths, London (1957).

Ridgway, K., and K. J. Tarbuck, "The Random Packing of Spheres," *Brit. Chem. Eng.*, **12**, 385 (1967).

Sareen, S. S., P. M. Rose, R. C. Gudeson, and R. C. Kintner, "Coalescence in Fibrous Beds," *AIChE J.*, **12** 1045 (1966).

Shalhoub, N., "Coalescence of Secondary Dispersions in Fibrous Beds," Ph.D. thesis, Univ. Aston, Birmingham, England (1975).

Spielman, L. A., and S. L. Goren, "Experiments in Coalescence by Flow through Fibrous Mats," *Ind. Eng. Chem. Fundamentals*, **11**, 73 (1972).

Spielman, L. A., and J. A. Fitzpatrick, "Theory for Particle Collection under London and Gravity Forces," *J. Coll. Interface Sci.*, **42**, 607 (1973).

Thomas, R. J., and C. J. Mumford, "Coalescence in Packed Beds—Packing Selection and Hydrodynamic Behaviour," *Proc. ISEC*, **1**, 400 (1971).

Wilkinson, D., "Studies of the Coalescence of Dispersions in Particulate packings," Ph.D. thesis, Univ. Aston, Birmingham, England (1974).

Manuscript received February 28, 1975; revision received May 12, and accepted May 13, 1975.

Self-Generated Oscillations in Continuous Crystallizers:

Part I. Analytical Prediction of the Oscillating Output

The theory of nonlinear oscillations should provide a useful tool for studying both forced and self-generated oscillations in continuous crystallizers. The application of the theory is described in this paper for the case of self-generated oscillations in a MSMPR (mixed-suspension, mixed product removal) isothermal crystallizer. Although the process parameters do not correspond to typical industrial conditions, the results indicate that for the Class I system studied higher yields sometimes can be obtained from the periodic process than the steady state design. Thus, there may be some situations where oscillatory operation is advantageous.

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SCOPE

Chemical processes, such as crystallizers, sometimes produce oscillating outputs, and in certain situations it may be desirable to predict the behavior of the oscillating system. In this paper it is shown that the theory of nonlinear oscillations gives reasonable analytical solutions for the periodic outputs. The theory is applied to the simplest kind of crystallizer model, that is, an isothermal, mixed suspension-mixed product removal unit for a Class I system where high degrees of supersaturation are possible. The focus of the study is on an evaluation of the method of analysis rather than the practical aspects of crystallization.

CONCLUSIONS AND SIGNIFICANCE

The results demonstrate that the theory of nonlinear oscillations provides an additional tool that can be used to study periodic operation of crystallizers. However, the theory is tedious to apply, so that future work should be directed toward the development of simpler approximate models. An interesting feature of the particular system studied was that the time-average yield of crystals produced by the oscillating system exceeded the predicted steady state value, although the design parameters considered were outside the normal range of industrial operating conditions.